PATENT
TS1175 (US)
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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF APPEALS AND INTERFERENCES

In re application of	)	
	)	
THEODORUS J. BROK, RUDOLF J. M. GROENEN, JEANINE M. KLINKENBIJL and MARIETTE C. KNAAP	)	
	)	
Serial No. 10/501,240	)	Group Art Unit: 1797
	)	
Filed March 30, 2005	)	Examiner: Ives J. Wu
	)	
PROCESS FOR REMOVING CARBON DIOXIDE FROM GAS MIXTURES	)	August 12, 2008
	)	

COMMISSIONER FOR PATENTS
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Sir:

APPEAL BRIEF

This brief is filed in support of Applicant's appeal from the Examiner's action dated March 13, 2008, finally rejecting claims 1-4, 9, 12-16, 20 and 23 of the above-identified U.S. patent application. A notice of appeal from this decision was filed by Applicant on June 16, 2008.

Please charge the fee for filing this brief to Shell Oil Company Deposit Account No. 19-1800.

It is respectfully requested that the Board reverse the final rejection of claims 1-4, 9, 12-16, 20 and 23 of the above-identified application for the reasons discussed below.

REAL PARTY IN INTEREST

The invention described and claimed in the above-identified patent application is assigned to Shell Oil Company, which is the real party in interest in the present appeal.

RELATED APPEALS AND INTERFERENCES

Appellant and Appellant's legal representatives are not aware of any related appeals or interferences.

STATUS OF CLAIMS

Claims 1-4, 9, 12-16, 20 and 23 have been rejected and are the subject of this appeal. A copy of the claims on appeal can be found in the Claims Appendix.

STATUS OF AMENDMENTS

Applicants' amendments filed January 7, 2008 were received and entered by the Examiner.

SUMMARY OF CLAIMED SUBJECT MATTER

The following is a summary of the claimed subject matter with specific reference to the portions of the specification that support the various claim limitations.

Independent claim 1 claims a process for removing carbon dioxide from a carbon dioxide-containing gas stream by washing the gas stream with an aqueous washing solution containing between 15 and 45 parts by weight water, between 15 and 40 parts by weight sulfolane and between 30 and 60 parts by weight of an amine selected from the group of amines consisting of MEA, DEA, TEA, DIPA and MDEA. The aqueous washing solution further contains from 0.7 mol/l to 0.9 mol/l of piperazine. Support for the limitations in claim 1 is found in the specification on page 3, line 20 to page 4, line 2, page 5, lines 9-22, page 6, line 31 to page 7, line 1 and page 1, line 28 to page 2, line 3.

Claim 15, the only other independent claim, claims an absorbent liquid having essentially the same composition as the aqueous washing solution used in the process of claim 1 (i.e., between 15 and 45 parts by weight water, between 15 and 40 parts by weight sulfolane and between 30 and 60 parts by weight of an amine selected from the group of amines consisting of

MEA, DEA, TEA, DIPA and MDEA, and from 0.7 mol/l to 0.9 mol/l piperazine). Support for the limitations in claim 15 is found in the specification on page 3, line 20 to page 4, line 2, page 5, lines 9-22, page 6, line 31 to page 7, line 1 and page 1, line 28 to page 2, line 3.

None of the dependent claims are being argued separately.

GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

The rejection of claims 1-4, 9, 12-16, 20 and 23 under 35 U.S.C. §102 (b) as being anticipated by Wagner et al (U.S. 4,997,630).

ARGUMENT

It is respectfully submitted that the rejection of claims 1-4, 9, 12-16, 20 and 23 under 35 U.S.C. §102 (b) as being anticipated by Wagner et al (U.S. 4,997,630) is erroneous and should be reversed.

The present invention is directed to process for removing carbon dioxide from streams containing high concentrations of carbon dioxide by use of a unique aqueous washing solution containing certain specified amounts of water, sulfolane (i.e., tetramethylenesulfone) and one of five specified amines, plus a specified concentration of piperazine. It has been found that contrary to teachings in the prior art, a mixture of water and a specific physical solvent, i.e., sulfolane, can be used at a relatively high concentrations in conjunction with piperazine and certain other amines without the formation of undesirable insoluble carbamates. (See page 3 of the specification, lines 5-19, wherein the teachings of U.S. 4,336,233 are discussed. A copy of U.S. 4,336,233 (which is referred to in the specification and thus is part of the record) is included in the Evidence appendix for the convenience of the Board.

In col. 3, lines 29-35 of U.S. 4,336,233 it is stated "that amongst industrially used physical solvents, e.g., methanol, mixtures of cyclotetramethylenesulfone, DIPA and water (Sulfinol®), NMP and dimethyl ethers of polyethylene glycols (Selexol®), only very dilute aqueous solutions can be used together with piperazine, because of the formation of piperazine carbamate." (emphasis added). Appellants' have surprisingly found that such limitations do not occur when mixtures of water/sulfolane in certain proportions are used with piperazine and MDEA.

The unique aqueous washing solution employed in the process recited in independent claim 1, and the unique absorbent liquid recited in independent claim 15 are specified to contain between 15 and 45 parts by weight water, between 15 and 40 parts by weight of sulfolane, between 30 and 60 parts by weight of an amine selected from the group consisting of MEA, DEA, TEA, DIPA and MDEA, and from 0.7 mol/l to 0.9 mol/l piperazine. The benefits of using this particular washing solution over prior art processes are discussed on page 4 of the specification lines 3-13. The benefits include faster carbon dioxide absorption rates resulting in higher CO_2 loadings, lower solvent/gas ratios allowing smaller plant size and lower regeneration heat requirement. The addition of sulfolane to the aqueous amine absorbent solution also allows the production of carbon dioxide at intermediate pressures, e.g. between 3 and 15 bara, preferably between 5 and 10 bara (specification, page 4, lines 13-16). Moreover, as previously discussed, the use of the specified amount of sulfolane as a physical solvent in the aqueous amine washing solution will not result in the formation of insoluble carbamates, which the prior art teaches can be a problem in aqueous amine systems containing piperazine, unless the physical solvents are used in very dilute aqueous solutions.

Appellant respectfully submits that the particular aqueous washing solution containing the specified concentrations of water, sulfolane, amine and piperazine recited in the present claims, is not anticipated by, nor would it be obvious from, Wagner et al for the reasons discussed below.

Wagner et al discloses a process for removing CO_2 and/or H_2S from gases using an aqueous alkanolamine-containing absorption liquid, wherein the gas is treated in a first absorption stage at 40° to 100° C, with an aqueous absorption liquid containing from 20 to 70 % by weight of methyldiethanolamine (MDEA). The gas obtained at the top of the first absorption stage is fed to a second absorption stage where it is treated at 30° to 90°C with an aqueous absorption liquid containing from 20 to 70 % MDEA to effect further removal of CO_2 and H_2S. The treated gas is taken off the top of the second absorption stage, while the aqueous absorption solution from the bottom of the second absorption stage preladen with CO_2 and/or H_2S is fed to the top of the first absorption stage. The aqueous absorption liquid obtained from the bottom of the first absorption stage laden with CO_2 and/or H_2S is let down in two or more flash stages in order to regenerate it (col. 1, lines 21-44).

In the sole example in the Wagner et al reference, a CO2-containing synthesis gas was washed with a 50% strength by weight aqueous methylenediethanolamine solution as the absorption liquid (col. 6, lines 56-59). This is the only specific absorption liquid disclosed in the Wagner et al reference. While the amine component (50 % strength by weight methylenediethanolamine) meets the 30 to 60 parts by weight amine limitation in the present claims, the absorption liquid used in the example does not contain sulfolane, does not contain piperazine and contains 50 % by weight water, which is outside the 15 to 45 parts by weight water limitation in the present claims.

Appellant acknowledges there are other disclosures in the Wagner et al reference on which the Examiner relies for his anticipation rejection. However, Appellant maintains these other disclosures do not teach the particular aqueous washing solution and absorbent liquid recited in the present claims with sufficient specificity to sustain an anticipation rejection. For example, the present claims require that the absorption solution contain between 15 and 45 parts by weight of a specific physical solvent, i.e., sulfolane. Wagner et al in its most relevant aspects teaches that the aqueous absorption liquid containing from 20 to 70 % by weight of methyldiethanolamine can additionally contain a physical solvent, and then goes on to list a number of physical solvents, including N-methylpyrrolidone, tetramethylene sulfone, methanol, oligoethylene glycol dialkyl ethers such as oligoethylene glycol methyl isopropyl ether (SEPASOLV MPE) or oligoethylene glycol dimethyl ether (SELEXOL) (Wagner et al, col. 2, lines 44-51). Wagner et al gives no specific examples where physical solvents are actually added to the absorption solution, and treats all of the listed physical solvents (if added at all) as being interchangeable.

In marked contrast, Appellant's aqueous washing solution must have a physical solvent and that physical solvent must be sulfolane. In addition, Appellant employs sulfolane at concentrations that one prior art patent teaches results in insoluble carbamate formation, which Appellant surprisingly found was not the case, as discussed above.

Another limitation in the present claims that Appellant respectfully submits is not anticipated by Wagner et al is the limitation that the aqueous wash solution contain between 15 and 45 parts by weight water. The only specific aqueous absorption liquid disclosed in Wagner et al is the absorption liquid used in the example in column 6 of the reference. That specific absorption liquid was 50 % by weight aqueous methyldiethanolamine solution. Since this

absorption liquid is described as a "50 % by weight aqueous methyldiethanolamine solution", it contains only two ingredients (50 %w methyldiethanolamine and water). Thus, the amount of water present in the absorption solution must also be 50 % by weight, which is outside of the range recited in the present claims.

In the final rejection the Examiner apparently recognizing that Wagner et al does disclose any specific compositions meeting the limitation that the aqueous washing solution contain between 15 and 45 parts by weight water, takes the position that "the balance of the aqueous washing liquid of Wagner et al (U.S. 4,997,630) would be water, which reads on the limitation as claimed". But to what "balance" does the Examiner refer? The balance of water in the aqueous washing liquid in the only example in Wagner et al is 50%, which is outside of the claimed range. There are three concentration ranges disclosed in Wagner et al for methyldiethanolamine (MDEA), i.e., 20 to 70 %w, 30 to 65 %w, and 40 to 60 %w. If a physical solvent is used at all, there are also three concentration ranges disclosed in Wagner et al for the physical solvent, i.e., 1 to 60 %w, 10 to 50 %w and 20 to 40 %w. Depending on how these ranges are combined, the "balance" of water in the absorption liquid of Wagner et al could range anywhere from a high of 79 %w if the absorption liquid contained the minimum amounts of MDEA and physical solvent (100% – (20% MDEA+1% physical solvent)), to a "balance" of water close to zero if the absorption liquid contained the maximum amounts of MDEA and physical solvent in the ranges specified by Wagner et al, e.g. 70 % MDEA and 60% physical solvent. Even if one used Wagner et al's preferred or most preferred ranges for MDEA and physical solvent, the "balance" of water would still vary widely, and at the most would only partially overlap the specific concentration range for water recited in the present claims. It is submitted that a partial overlap of a specific range does not anticipate the specific range, especially in a case such as this where the only actual example in the reference falls outside of the claimed range.

SUMMARY

Wagner et al does not anticipate the subject matter claimed in present claims in that Wagner et al does not disclose an aqueous washing solution containing between 15 and 45 parts by weight water, between 15 and 40 parts by weight of sulfolane, between 30 and 60 parts by weight of an amine selected from the group consisting of MEA, DEA, TEA, DIPA and MDEA, and from 0.7 mol/l to 0.9 mol/l piperazine.

More specifically, Wagner et al does not anticipate the limitation that the aqueous washing solution must contain between 15 and 40 parts by weight sulfolane. The Wagner et al reference only discloses that a physical solvent can be added to the aqueous absorption liquid used in their two stage absorption process. There is no requirement that a physical solvent actually be added. Moreover, Wagner et al only discloses sulfolane as one of a number of physical solvents that can be used. Furthermore, the only specific absorption liquid disclosed by Wagner et al (the absorption liquid in the example), does not contain sulfolane or any other physical solvent.

In addition, Wagner et al does not anticipate the limitation that the aqueous wash solution contain between 15 and 45 parts by weight water. The only specific absorption liquid disclosed in Wagner et al contains 50 %w of water, which is outside of the range specified in the present claims. The Examiner's position that the "balance" of the aqueous washing liquid in Wagner et al would read on the between 15 and 45 parts by weight water limitation is untenable, because Wagner et al discloses multiple concentration ranges for MDEA and multiple concentration ranges for the physical solvent (if a physical solvent is even used). Under these circumstances it is not possible to determine the "balance" of water present in the aqueous absorption liquid with any specificity, since the concentration of the MDEA and physical solvent, if present at all, can vary widely.

Therefore, Applicant's discovery of a unique and highly effective aqueous washing solution having the particular specified concentrations of four particular required ingredients, one of which must be sulfolane, to treat gas streams having a high concentration of carbon dioxide is not anticipated by Wagner et al.

While the present claims are clearly not anticipated by Wagner et al, which is the basis for the rejection being appealed, it is noted for the sake of record that the present claims are also believed to be unobvious over Wagner et al. This belief is based on the fact that the prior art, such as U.S. 4,336,233, discussed in the specification, teaches that if physical solvents are employed in aqueous amine solutions containing piperazine, they should be employed only in very dilute aqueous solutions. Applicants have surprisingly found that this is not true in case of at least one specific physical solvent, i.e., sulfolane, which is employed in Applicants' aqueous washing solution in the relatively high concentration of 15 to 40 parts by weight. It is respectfully submitted that an aqueous washing solution having the specified concentration of sulfolane and

the specified concentrations of the other ingredients recited in the present claims, is not obvious from the prior art taken as a whole.

CONCLUSION

For all the above stated reasons, independent claims 1 and 15, and the remaining claims that are directly or indirectly dependent thereon and therefore contain similar limitations, are believed to be patentable over the cited reference. Accordingly, the action of the Examiner in finally rejecting these claims should be reversed, which action is respectfully requested.

Respectfully submitted,

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CLAIMS APPENDIX

1. A process for the removal of carbon dioxide from a gas stream containing carbon dioxide by washing the gas stream with an aqueous washing solution containing between 15 and 45 parts by weight water, between 15 and 40 parts by weight sulfolane and between 30 and 60 parts by weight of an amine selected from the group of amines consisting of MEA, DEA, TEA, DIPA and MDEA, wherein the parts by weights are based on the amounts of water, sulfolane and amine together being 100 parts by weight, and, further, wherein the aqueous washing solution contains piperazine in an amount in the range of from 0.7 mol/l to 0.9 mol/l.

2. The process of claim 1, wherein the gas stream is natural gas or synthesis gas.

3. The process of claim 2, wherein the gas stream includes an amount of carbon dioxide that is between 1 and 45 mol%, an amount of hydrogen sulphide that is between 0 and 25 mol%, and an amount of COS that is between 0 and 2 mol% (all % based on total gas stream).

4. The process of claim 3, wherein the aqueous washing solution includes an amount of water that is between 20 and 45 parts by weight, an amount of sulfolane that is between 20 and 35 parts by weight, and an amount of the amine that is between 40 and 55 parts by weight, wherein the parts by weights are based on the amounts of water, sulfolane and amine together being 100 parts by weight.

9. The process of claim 4, wherein the piperazine is present in the aqueous washing solution in an amount in the range of from 0.6 to 0.8 mol/l.

12. The process of claim 9 wherein the process is carried out at a temperature of at least 20°C.

13. The process of claim 12, wherein the process also comprises a regeneration of the loaded solvent.

14. The process of claim 13, wherein the process is carried out at a pressure between 25 and 90 bara.

15. An absorbent liquid containing between 15 and 45 parts by weight water, between 15 and 40 parts by weight sulfolane and between 30 and 60 parts by weight of an amine selected from the group of amines consisting of MEA, DEA, TEA, DIPA and MDEA, wherein the parts by weights are based on the amounts of water, sulfolane and amine together being 100 parts by weight, and, further, wherein the aqueous washing solution contains piperazine in an amount in the range of from 0.7 mol/l to 0.9 mol./l.

16. The absorbent liquid as defined in claim 15, wherein the amount of water is between 20 and 45 parts by weight, the amount of sulfolane is between 20 and 35 parts by weight, and the amount of amine is between 40 and 55 parts by weight, wherein the parts by weights are based on the amounts of water, sulfolane and amine together being 100 parts by weight.

20. The absorbent liquid of claim 16, wherein the piperazine is present in the aqueous washing solution in an amount in the range of from 0.6 to 0.8 mol/l.

23. The absorbent liquid of claim 20, wherein the amine is MDEA.

EVIDENCE APPENDIX

This Appendix contains a copy of U.S. 4,336,233 to Appl et al, which is referred to on page 3 of the present application.

RELATED PROCEEDINGS APPENDIX

None